

Scalings and Limits of the Landau-de Gennes Model for Liquid Crystals

A Comment on Some Recent Analytical Papers

Eugene C. Gartland, Jr.

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Abstract Recent analytical papers in this journal have explored limiting behaviors of the Landau-de Gennes model for liquid crystals in certain extreme ranges of the model parameters. We use a simple scaling analysis to show that these limits are properly interpreted as limits in which geometric length scales (such as the size of the domain containing the liquid crystal material) become large compared to intrinsic length scales (such as defect core sizes). This represents the natural passage from a mesoscopic model to a macroscopic model and is analogous to a “London limit” in the Ginzburg-Landau theory of superconductivity.

Keywords Liquid Crystals · Landau-de Gennes Model · Oseen-Frank Model

1 Introduction

The Landau-de Gennes and Oseen-Frank models are the two most widely used continuum models to characterize equilibrium orientational properties of materials in the nematic liquid crystal phase. The Landau-de Gennes model is a mesoscopic phenomenological model expressed in terms of an integral functional of a tensor field Q , the “tensor order parameter.” In its simplest form (the “equal elastic constant model”), the functional can be written

$$\mathcal{F}[Q] = \int_{\Omega} \left[\frac{L}{2} |\nabla Q|^2 + f_b(Q) \right] dV. \quad (1a)$$

Here \mathcal{F} gives the free energy of a material occupying the region Ω , and f_b represents the bulk free energy density

$$f_b(Q) = \frac{A}{2} \text{tr}(Q^2) - \frac{B}{3} \text{tr}(Q^3) + \frac{C}{4} \text{tr}(Q^2)^2. \quad (1b)$$

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E. C. Gartland, Jr.
Department of Mathematical Sciences, Kent State University, Kent, Ohio 44242, USA
Tel.: +1-330-672-9112, Fax: +1-330-672-2209, E-mail: gartland@math.kent.edu

The parameters L , A , B , and C are material dependent, with L , B , and C positive and A (which can be positive, negative, or zero) usually taken to have a simple linear dependence on temperature:

$$A = a(T - T_{\text{SC}}), \quad a > 0.$$

Here T denotes temperature and T_{SC} represents the “super-cooling temperature” (discussed below). Stationary points of \mathcal{F} give equilibrium orientational states, with the global minimum determining the phase of the system for a given set of boundary conditions and parameter values.

The \mathbf{Q} tensor is formally defined as the traceless part of the second-moment tensor of the orientational probability distribution function (which, if known, would contain complete information about the orientational state at a point):

$$\mathbf{Q} = \langle \mathbf{l} \otimes \mathbf{l} \rangle - \frac{1}{3} \mathbb{I}.$$

Here the unit vector \mathbf{l} denotes the direction of the distinguished axis of the anisometric molecular architecture (usually the long axis of elongated, rod-like molecules) and \mathbb{I} is the identity tensor. As such, \mathbf{Q} is real, symmetric, traceless, and generically would have three distinct real eigenvalues and associated orthogonal eigenvectors, representing a “biaxial” state of order. The eigenvectors of \mathbf{Q} provide information about the directions of orientational ordering at a point, while the eigenvalues give information about the degrees of order—note that f_b depends only on the eigenvalues of \mathbf{Q} . An isotropic (totally disordered) state corresponds to $\mathbf{Q} = 0$ (the zero tensor). For given $B, C > 0$, the shape of the bulk free energy surface of f_b , as a function of the two independent eigenvalues of \mathbf{Q} , changes as temperature changes. For $T > T_{\text{SH}} > T_{\text{SC}}$, where T_{SH} denotes the “super-heating temperature,” $\mathbf{Q} = 0$ is the global minimizer of f_b , and no ordered phase exists— f_b has no other stationary points. For $T < T_{\text{SC}}$, the isotropic phase ceases to be locally stable, and the only minimizer of f_b is a \mathbf{Q} tensor with a dominant eigenvalue and a degenerate pair of equal eigenvalues. Since $\text{tr}(\mathbf{Q}) = 0$, such a \mathbf{Q} can be written

$$\mathbf{Q} = S \left(\mathbf{n} \otimes \mathbf{n} - \frac{1}{3} \mathbb{I} \right), \quad (2)$$

where S is the dominant eigenvalue (the “scalar order parameter”) and \mathbf{n} its associated eigenvector. This is referred to as a “uniaxial” state of order. For T in the narrow range between T_{SC} and T_{SH} , both the isotropic and the uniaxial phases are locally stable, and there is a first-order phase transition at the “nematic-isotropic transition temperature” T_{NI} , below which the ordered phase becomes the global minimizer. As T decreases below this point, the degree of order S increases. Simple relationships among A , B , and C are associated with these temperatures:

$$T_{\text{SC}} < T_{\text{NI}} < T_{\text{SH}} \quad \leftrightarrow \quad A_{\text{SC}} = 0 < A_{\text{NI}} = \frac{B^2}{27C} < A_{\text{SH}} = \frac{B^2}{24C}. \quad (3)$$

The distortional elastic term associated with the elastic constant L in (1a) penalizes spatial variations of the \mathbf{Q} tensor, while the bulk ordering potential f_b strives to

put the eigenvalues of Q into certain wells. More complete versions of this model can contain additional terms incorporating more elastic constants, more bulk constants, terms associated with couplings to electric or magnetic fields, terms associated with chirality, flexoelectric effects, ferroelectric effects, surface anchoring potentials, and more. See [15] for a gentle introduction to the model or [6, 22] for textbook treatments. The form (1) is the simplest form of the model and is sufficient for the purposes at hand. Landau-de Gennes models occupy a similar position in the theory of liquid crystals to that of Ginzburg-Landau models for the theory of superconductivity.

The Oseen-Frank model is a macroscopic phenomenological model for liquid crystal orientational properties. It is expressed in terms of an integral functional of a unit-length vector field \mathbf{n} (the “director field”). In its simplest (“equal elastic constant”) form, the functional can be written

$$F[\mathbf{n}] = \frac{K}{2} \int_{\Omega} |\nabla \mathbf{n}|^2 dV.$$

Here F gives the distortional elastic energy of the material occupying Ω , and \mathbf{n} represents the average orientation of the distinguished axis of the molecules in a fluid element at a point (and can be identified with the eigenvector associated with the distinguished eigenvalue of a uniaxial Q tensor, as in (2)). The elastic constant K is a material-dependent and temperature-dependent parameter. Equilibrium orientational states are given by stationary points of F constrained by boundary conditions and the pointwise unit-length constraint on \mathbf{n} , with the phase of the system (for a given K and boundary conditions) again given by the global minimizer. As is the case with the Landau-de Gennes free energy \mathcal{F} , more realistic models for F involve many more terms, parameters, couplings, and effects. Standard references include [4, 6, 23, 24]. Oseen-Frank models have been widely and successfully used for many years to model liquid crystal systems at typical device and experimental scales.

These two models differ in several ways. The Landau-de Gennes model allows for both spatially varying degrees of order and “biaxiality,” and it is expressed in terms of a tensor field. The Oseen-Frank model, on the other hand, assumes a uniform degree of orientational order, as well as a “uniaxial” state of order, and is expressed in terms of a vector field. Another difference between the models is that common liquid crystal “defects” (such as “point defects” and “disclination lines”) are singularities of the director field in the Oseen-Frank model, whereas in the Landau-de Gennes model, these have a small but finite “core size” and the associated tensor field Q remains smooth throughout a neighborhood of such a defect. In the Oseen-Frank model, point defects have finite free energy, while the free energy of disclinations is infinite. In the Landau-de Gennes model, however, the free energies of both are finite. In general, defects are caused by the conflicting demands of distortional elasticity, boundary conditions, external electric or magnetic fields, and the like. The Landau-de Gennes model is typically employed in problems in which geometric length scales are not too large compared to intrinsic length scales (core sizes), while the Oseen-Frank model is used when the geometric length scale is much larger than the core size (and the fine-detail structure of the core is not important).

In recent years, the Landau-de Gennes model has received considerable attention from the mathematical analysis community. In particular, three papers have appeared

in this journal that rigorously explore (among other issues) limiting behaviors of this model for certain extreme ranges of parameter values [1, 11, 14]. Our main purpose here is to explain how these limits should be interpreted.

In [14] (the earliest of these papers), the model is taken in the form (1) above, and the authors motivate their work as follows: “we study the limit of vanishing elastic constant $L \rightarrow 0 \dots$ the limit $L \rightarrow 0$ is a physically relevant limit since the elastic constant L is typically very small, of the order 10^{-11} J/m.” In [1], a slightly more general form of the model is used:

$$\mathcal{F}_\varepsilon[\mathbf{Q}] = \int_\Omega [f_e(\nabla \mathbf{Q}) + \varepsilon^{-2} f_b(\mathbf{Q})] dV, \quad (4a)$$

where

$$f_e(\nabla \mathbf{Q}) = \frac{L_1}{2} Q_{ij,k} Q_{ij,k} + \frac{L_2}{2} Q_{ij,j} Q_{ik,k} + \frac{L_3}{2} Q_{ij,k} Q_{ik,j}. \quad (4b)$$

Here Q_{ij} are the components of \mathbf{Q} with respect to a fixed Cartesian frame, and the dimensionless parameter ε is artificially introduced in order to be able to drive L_1, L_2 , and L_3 to zero simultaneously. The L_1 term above corresponds to the L term in (1a). The authors motivate their work “Our goal in this paper is to investigate minimizers [of (4)] and to analyze their behavior in the vanishing elastic energy limit, $\varepsilon \rightarrow 0$.”

In these two papers, the models are analyzed in fully dimensional form. In [11], in contrast, a dimensionless model is studied, and it is expressed in terms of the second-moment tensor $\mathbf{u} = \langle \mathbf{I} \otimes \mathbf{I} \rangle$ (instead of \mathbf{Q}). The model takes the form

$$E_\varepsilon[\mathbf{u}] = \int_\Omega \left[\frac{|\nabla \mathbf{u}|^2}{2} + \frac{W(\mathbf{u})}{\varepsilon^2} \right] dV, \quad (5a)$$

where

$$W(\mathbf{u}) = \frac{1}{2} \text{tr}((\mathbf{u} - \mathbf{u}^2)^2). \quad (5b)$$

The bulk ordering potential W here is constructed by design to have a minimizer (in the class $\text{tr}(\mathbf{u}) = 1$) at a perfectly ordered uniaxial state $\mathbf{u} = \mathbf{n} \otimes \mathbf{n}$, with $|\mathbf{n}| = 1$ but otherwise arbitrary, and ε is referred to as a “dimensionless elastic constant.” With the help of the relation $\mathbf{u} = \mathbf{Q} + \frac{1}{3} \mathbb{I}$, the model can be identified with a certain constrained form of (1). Again, the limit $\varepsilon \rightarrow 0$ is explored.

All three papers explore similar limits, related to “vanishing elasticity,” and are influenced by similar analyses of Ginzburg-Landau models found in [2, 3] and elsewhere. All three obtain (away from a singular set) limiting uniaxial minimizers of the form (2), with constant S determined by f_b , and with the director field \mathbf{n} corresponding to the minimizer of an appropriate Oseen-Frank model. We show that these limits are properly interpreted not as limits of vanishing elasticity but as limits in which intrinsic length scales (associated with defect core sizes and such) become vanishingly small compared to geometric length scales (associated with the size of the problem domain Ω).

2 Analysis

Values of the material parameters in (1) are usually found quoted in SI units. For a somewhat typical material, they are roughly in the following ranges:

$$L \approx 10^{-11} \text{J/m}, \quad A, B, C \approx 10^3 \text{J/m}^3.$$

See for example [18, Table 1, p. 168]. We observe that the numerical value of L is 14 orders of magnitude smaller than the values of A , B , and C when expressed in these units. If, however, lengths are expressed in units of nanometers (instead of meters), these values become

$$L \approx 10^{-20} \text{J/nm}, \quad A, B, C \approx 10^{-24} \text{J/nm}^3.$$

Now the value of L is four orders of magnitude *larger* than A , B , and C . The point is that the elastic constants and the bulk constants have different physical dimensions (energy per unit length versus energy per unit volume) and can't be compared. To determine what is "big" versus what is "small," one must non-dimensionalize. An additional point to be made is that as $L \rightarrow 0$, with the model left in dimensional form (as in [1] and [14]), one quickly gets beyond the measured values of L for real liquid crystal materials.

There are numerous ways to non-dimensionalize (1), and these depend in general on the particular problem at hand. In essence, in order to form an appropriate dimensionless coupling coefficient between the elastic terms and the bulk terms in the free-energy density, one requires a characteristic elastic constant, a characteristic bulk constant, and a characteristic length scale from the geometry of the problem domain. For our purposes here, a simple rescaling can be done as follows. Let R denote a characteristic geometric length scale. For example, this could be the radius or diameter of a liquid crystal droplet or capillary, or the cell gap of a liquid crystal thin film. Rescale lengths by R ,

$$\bar{x}_i = \frac{x_i}{R}, \quad R = \text{diam}(\Omega),$$

so that

$$\nabla = \frac{1}{R} \bar{\nabla}, \quad dV = R^3 d\bar{V}, \quad \text{diam}(\bar{\Omega}) = 1.$$

Let A_{NI} denote the value of the A parameter at $T = T_{\text{NI}}$ and take this as our characteristic bulk parameter—other natural candidates for this would be A_{SC} or simply B^2/C . The tensor order parameter Q is dimensionless by definition, however it is convenient to rescale it as well, as this allows one to eliminate another parameter from the model. Thus we take

$$Q = \alpha \bar{Q}, \quad \alpha = \sqrt{\frac{2}{27} \frac{B}{C}}.$$

Other multiples of B/C would work equally well. After simplifying, we obtain

$$\mathcal{F}[\bar{Q}] = \int_{\bar{\Omega}} \left[\frac{\varepsilon^2}{2} |\bar{\nabla} \bar{Q}|^2 + \frac{\theta}{2} \text{tr}(\bar{Q}^2) - \sqrt{6} \text{tr}(\bar{Q}^3) + \frac{1}{2} \text{tr}(\bar{Q}^2)^2 \right] d\bar{V}, \quad (6a)$$

where

$$\overline{\mathcal{F}} = \frac{\mathcal{F}}{\alpha^2 A_{\text{NI}} R^3}, \quad \varepsilon = \frac{\xi}{R}, \quad \xi = \sqrt{\frac{L}{A_{\text{NI}}}}, \quad \theta = \frac{A}{A_{\text{NI}}} = \frac{T - T_{\text{SC}}}{T_{\text{NI}} - T_{\text{SC}}}. \quad (6b)$$

The parameter ξ can be interpreted as a “nematic correlation length” and θ as a reduced temperature, with the corresponding values

$$T = T_{\text{SC}}, T_{\text{NI}}, T_{\text{SH}} \leftrightarrow \theta = 0, 1, 9/8.$$

Here and in (6), we have used the relations for A_{NI} and A_{SH} in (3). In a typical liquid crystal system (for example, a thin film of cell gap R containing a low-molecular-weight liquid crystal material), we could have

$$\xi \approx 10 \text{ nm}, \quad R \approx 10 \mu\text{m} \Rightarrow \varepsilon = \frac{\xi}{R} \approx 10^{-3}.$$

The nematic correlation length has a statistical physics interpretation, however in our continuum model, it simply emerges as a singular perturbation parameter in the Euler-Lagrange equations associated with (1):

$$-L\Delta Q + \overline{\frac{\partial f_b}{\partial Q}} = 0, \quad (7)$$

where $\overline{\cdot}$ denotes the symmetric traceless part:

$$\overline{\frac{\partial f_b}{\partial Q}} = A Q - B \left(Q^2 - \frac{1}{3} \text{tr}(Q^2) \mathbb{I} \right) + C \text{tr}(Q^2) Q.$$

At $T = T_{\text{NI}}$, we have

$$-\xi^2 \Delta Q + \frac{1}{A_{\text{NI}}} \overline{\frac{\partial f_b}{\partial Q}} = 0.$$

When Q is close to a stationary point of f_b (such as $Q = 0$ or the bottom of a well), then the term $(1/A_{\text{NI}}) \overline{\partial f_b / \partial Q}$ will be close to zero; otherwise, it will be $O(1)$. The necessary balance between the terms in the equation above indicates how ξ essentially determines the “core size” of a defect.

We have defined ξ by linearization around the isotropic state $Q = 0$, which is always a critical point of f_b and the solution one would see at the center of an isotropic core—note however that the core structure of a defect is generally more complicated than isotropic disordering (see [20]). In some settings, it is more natural to define ξ as the coupling coefficient between the two terms in (7) when linearized around the nontrivial uniaxial Q tensor that gives the global minimum of f_b for $T < T_{\text{NI}}$ —see for example [19]. Core sizes depend on temperature, and this should be taken into account in non-dimensionalizations if one intends to explore behavior deep in the nematic phase. In some situations, a more significant intrinsic length scale is given by a “biaxial coherence length”—see [12] or [13]. The scalings we have adopted here have been chosen for simplicity. They are by no means original. Similar scalings are employed by all practitioners who work with Landau-de Gennes models. In addition to references already cited, see for example [7, 8, 9, 10, 17, 21, 25], among others.

In our rescaled free energy (6), all quantities are dimensionless, and $\overline{\mathcal{F}}$, $\overline{\mathbf{Q}}$, θ , and the size of $\overline{\Omega}$ are $O(1)$. We can see under what circumstances the coupling coefficient between the elastic and bulk terms is small:

$$\varepsilon \ll 1 \Leftrightarrow \xi \ll R,$$

that is, when the nematic correlation length (core size) is small compared to a length scale associated with the problem geometry (size of Ω). The dimensionless spatial gradient $\nabla \overline{\mathbf{Q}}$ will be $O(1)$ away from defects and $O(1/\varepsilon)$ in the vicinity of defects. The limit $\varepsilon \rightarrow 0$ corresponds to the core size of defects becoming vanishingly small compared to the size of the problem geometry, with finite-size defects becoming point or line singularities in the limit. We think of this as a “zero-core-size limit.”

For the more general functional (4), a similar rescaling would give

$$\begin{aligned} \overline{\mathcal{F}}_\varepsilon[\overline{\mathbf{Q}}] = \int_{\overline{\Omega}} \bigg[\frac{\varepsilon^2}{2} \big(\overline{\mathbf{Q}}_{ij,\bar{k}} \overline{\mathbf{Q}}_{ij,\bar{k}} + \overline{L}_2 \overline{\mathbf{Q}}_{ij,\bar{j}} \overline{\mathbf{Q}}_{ik,\bar{k}} + \overline{L}_3 \overline{\mathbf{Q}}_{ij,\bar{k}} \overline{\mathbf{Q}}_{ik,\bar{j}} \big) \\ + \frac{\theta}{2} \text{tr}(\overline{\mathbf{Q}}^2) - \sqrt{6} \text{tr}(\overline{\mathbf{Q}}^3) + \frac{1}{2} \text{tr}(\overline{\mathbf{Q}}^2)^2 \bigg] d\overline{V}, \end{aligned} \quad (8a)$$

where

$$\varepsilon = \frac{\xi}{R}, \quad \xi = \sqrt{\frac{L_1}{A_{\text{NI}}}}, \quad \overline{L}_2 = \frac{L_2}{L_1}, \quad \overline{L}_3 = \frac{L_3}{L_1}. \quad (8b)$$

Here \overline{L}_2 and \overline{L}_3 are dimensionless and $O(1)$. The definition of the dimensionless parameter ε in (5a) is not given in [11]. One can assume that it has been constructed in a way that is analogous to what has been done here for ε in (6) and above.

3 Conclusions

The Landau-de Gennes model is a mesoscopic model that contains intrinsic length scales of molecular order associated with features such as core sizes of point defects and disclination lines. The Oseen-Frank model, on the other hand, is a macroscopic model and contains no such intrinsic length scales. The defects of equilibrium director fields of the Oseen-Frank model are point or line singularities. The analysis of the preceding section shows that the limits explored in [1, 11, 14] concern the passage from a mesoscopic model to a macroscopic model as the geometric length scales become large compared to the intrinsic length scales. In such a limit, core sizes become zero. The situation is analogous to a “London limit” for Ginzburg-Landau models in which the role of the Ginzburg-Landau parameter is here played by the ratio of an intrinsic length scale to a geometric one—a proper scaling and non-dimensionalization of the model are required to identify this. The limit (properly interpreted) can indeed be thought of as “the Oseen-Frank limit,” as in the title of [14].

The analyses in these papers remains valid, once a re-interpretation is done of the problem parameters (into their appropriate dimensionless forms)—the functionals (1a) and (6a) involve the same terms in a formal sense, just differently scaled. One should take heed, however, of the pitfalls of analyzing such a physical model in fully dimensional form. The numerical values of quantities of different physical

dimensions can change their relative sizes when the system of units is changed. It is also the case that familiar Sobolev-type norms, such as

$$\|Q\|_1^2 = \int_{\Omega} [|\nabla Q|^2 + |Q|^2] dV$$

(which are used in most of the papers in this area), can't even be used in this form unless lengths have been non-dimensionalized; otherwise the first term in the integrand above would have dimensions of $1/\text{length}^2$, while the second term would be dimensionless (and the two terms couldn't be combined). An alternative remedy for this would be to use weighted Sobolev norms, such as

$$\|Q\|_1^2 = \int_{\Omega} [w_1 |\nabla Q|^2 + w_2 |Q|^2] dV,$$

with w_1 and w_2 chosen with dimensions to render the combination meaningful. We acknowledge that [5] is guilty of this error as well.

An additional difficulty of attempting to analyze such limits in dimensional form is that as $L \rightarrow 0$ in (1a), one quickly gets beyond the measured values of L for real liquid crystal materials (as has already been observed). A properly non-dimensionalized model such as (6), however, does not suffer from this difficulty: the smallness of the coupling coefficient ε^2 in (6a) or (8a) is related only to the ratio of the core size to the size of the problem domain.

While the analyses in [1, 11, 14] are modeled after earlier work by others on models with some similar features (such as Ginzburg-Landau and Chern-Simons-Higgs), technical challenges accompany the analysis of Landau-de Gennes models by virtue of the tensorial nature of the state variable and the multiple terms and parameters and the complexity of the functional. The simple scaling analysis done here shows that the limits explored in these papers are properly interpreted as limits in which the size of the problem domain becomes large compared to the core size of defects, a natural passage from a mesoscopic model to a macroscopic one. An effort to address some of the issues taken up in this note is made in a brief appendix in [16], which seems to have been otherwise overlooked.

One of the interesting aspects of the asymptotic analyses done in the papers [1, 11, 14] is that they provide, to some extent, a justification for using the Oseen-Frank model to compute equilibrium director fields in the case when line disclinations are present (in the regime of "large" physical domains): even though the Oseen-Frank free energy of such solutions is infinite, the equilibrium director fields (found from the Euler-Lagrange equations) are the limiting director fields associated with solutions of the Landau-de Gennes model, which solutions have finite free energy for all positive values of the coupling coefficient ε , though diverging like $|\ln \varepsilon|$ in the limit as $\varepsilon \rightarrow 0$. The divergence of the Oseen-Frank free energy F , however, prevents it from being used to assess local or global stability of such solutions.

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